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AGRICULTURE AND GARDEN MICROBICIDE COMPOSITION

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AGRICULTURE AND GARDEN MICROBICIDE COMPOSITION

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Claim

An agriculture and garden microbicide composition characterized by comprising amido derivatives represented by general formula

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\* [Numbers in margin indicate pagination in the foreign text.]



//insert formula marked "A" on page 13//

[wherein R<sup>1</sup> represents methyl group or ethyl group, and X represents oxygen atom or sulfur atom] and imidazole derivatives represented by general formula

//insert formula marked "B" on page 13//

[wherein R<sup>2</sup> represents alkyl group or phenyl group optionally having halogen substituents, and Y represents halogen atom], as the active ingredients.

#### Detailed explanation of the invention

##### Industrial application field

This invention pertains to an agriculture and garden microbicide composition.

##### Prior art

Japanese Kokai Patent Application No. Hei 1[1989]-301681 disclosed the effectiveness of certain amido derivatives as agriculture and garden microbicide. Additionally, Japanese Kokai Patent Application No. Hei 1[1989]-131163 disclosed the effectiveness of certain imidazole derivatives as agriculture and garden microbicide.

##### Problem to be solved by the invention

However, the potencies of these agriculture and garden microbicides are not necessarily considered sufficient.



Means to solve the problem

Focusing on such current conditions, the present inventors realized the necessity of developing compositions having excellent potency as agriculture and garden microbicides and conducted various investigations and, as a result, discovered that, when combining certain kinds of imido derivatives and certain kinds of imidazole derivatives, unexpectedly excellent microbicide potency can be realized compared to when each kind of compound is utilized alone. /2

That is, the present invention is to provide an agriculture and garden microbicide composition (hereafter as "composition of the present invention" characterized by comprising amido derivatives represented by general formula

//insert formula I on page 14//

[wherein  $R^1$  represents methyl group or ethyl group, and X represents oxygen atom or sulfur atom] and imidazole derivatives represented by general formula

//insert formula II on page 14//

[wherein  $R^2$  represents alkyl group or phenyl group optionally having halogen substituents, and Y represents halogen atom], as the active ingredients.

Table 1 shows the examples of amido derivatives represented by general formula (I) that are utilized in the present invention.



Table 1

//insert Table, page 14//

Key	1	Compound symbol
	2	Chemical structure



Table 2 shows the examples of imidazole derivatives represented by general formula (II) that are utilized in the present invention.

Table 2

//insert Table 2 on page 14//

Key	1	Compound symbol
	2	Chemical structure

The examples of the plant diseases that the composition of the present invention is capable of eliminating are diseases of phycomycetes that are cited in the following.

*Peronospora brassicae* of the vegetables and daikon family, *Peronospora spinaciae* of spinach, *Peronospora tabacina* of tobacco, *PseudoPeronospora cubensis* of melon, *Plasmopara viticola* of grape, *Phytophthora cactorum* of apple, strawberry, medicinal carrot, *Phytophthora*



*capsici* of tomato and cucumber, *Phytophthora cinnamomi* of pineapple, *Phytophthora infestans* of potato, tomato and eggplant, *Phytophthora nicotianae* var. *nicotianae* of tobacco, broad bean and scallion, *Pythium* sp. of spinach, *Pythium aphanidermatum* of cucumber, *Pythium debaryanum* of tobacco, *Pythium* sp. of wheat, and *Pythium* rot (*Pythium irregulare aphanidermatum*, *P. debaryanum*, *P. irregulare*, *P. myiotylum* [sic; *myriotylum*], *P. ultimam*) of soybean.

Therefore, the composition of the present invention can be applied to farming fields and fruit farms as the active ingredient of a microbicide.

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If the composition of the present invention is utilized as the active ingredient of a microbicide, it can be used as it is without adding any other ingredient, but in general, a solid carrier, liquid carrier, surfactant and other supplementary agents for preparations are mixed to make preparations as aqueous, suspension, granular, powder or microgranular agents.

As examples of the solid carriers that can be utilized, powder or granules of kaolin clay, attapulgit clay, bentonite, acidic clay, pyrophyllite, talc, diatomaceous earth, calcite, walnut shell powder, urea, ammonium sulfate, and micropowders or granulates of synthetic hydrated silicon oxide can be cited.

As the examples of the liquid carrier that can be utilized, aromatic hydrocarbons such as xylene and methylnaphthalene, alcohols such as isopropanol, ethylene glycol and cellosolve, ketones such as acetone, cyclohexane [sic; cyclohexanone] and isophorone, vegetable oils such as soybean oil and cottonseed oil and dimethylsulfoxide, acetonitrile and water can be cited.

As the examples of the surfactants utilized for emulsification, dispersion and wet spreading, anionic surfactants such as alkyl sulfate salts, alkyl (aryl) sulfonic acid salts, dialkyl sulfosuccinic acid, polyoxyethylene alkylaryl ether phosphate salts and condensation products of naphthalene sulfonic acid-formalin, and nonionic surfactants such as polyoxyethylene alkyl ether, polyoxyethylene alkylaryl ether, polyoxyethylene polyoxypropylene block copolymers, sorbitan fatty acid esters, and polyoxyethylene sorbitan fatty acid esters can be cited.

As the examples of the supplementary agents for the preparations, lignin sulfonic acid



salts, alginic acid salts, polyvinyl alcohol, gum arabic, CMC (carboxymethylcellulose) and PAP (isopropyl acid phosphate) can be cited.

The mixing ratio of the active ingredients of the present invention, amido derivative (I) and imidazole derivative (II), in general is 1:5 to 5:1 by weight, and the total amount of the active ingredients in the aforementioned preparations in general is 0.1-99.9%, preferably 0.2-80% by weight.

The aforementioned preparations may be applied in many forms, including spraying onto the stems and leaves as they are or after diluting with water, or dispersing and mixing as powder or granulates in the soil, or just simply applying to the soil. Also, enhancement of the prevention and elimination effect can be expected by mixing with other microbicides. Furthermore, They can be used together with insecticides, miticides, nematocides, weed killers, plant growth regulating agents, fertilizers and soil improving agents.

If the composition of the present invention is applied as a plant disease prevention agent, a suitable amount of the active ingredient to be applied is generally 0.01-50 g per are, preferably 0.05-10 g per are; if it is applied by diluting with water including hydrating, suspending agent and emulsifying agents, the preferred amount of application is 0.001-0.2%. Also, in the case of powder or granular agent, it is applied as is without diluting. The amount of application or concentration of application of these forms depend on the form of the preparations, time of application, location, method of application, type and degree of plant disease and the type of crop, and they can be increased or decreased without being restricted by the aforementioned ranges.

#### Application examples

The present invention is further explained in detail below using manufacturing examples and test examples, but the application examples are not to be construed as limiting the present invention.

First, manufacturing examples are shown. Here, the parts are shown in parts by weight.



Manufacturing Example 1 Powder agent

One part each of compounds Ia-Id, 1 part each of compounds IIa-IIc, 88 parts kaolin clay and 10 parts talc were thoroughly pulverized and mixed to give a 2% powder agent.

Manufacturing Example 2 Hydrated agent

Three parts each of compounds Ia-Id, 15 parts each of compounds IIa-IIc, 57 parts diatomaceous earth, 20 parts white carbon, 3 parts wetting agent (sodium lauryl sulfate) and 2 parts dispersing agent (calcium lignin sulfonate) were thoroughly pulverized and mixed to give a 18% hydrated agent. /4

Manufacturing Example 3 Hydrated agent

Fifteen parts each of compounds Ia-Id, 3 parts each of compounds IIa-IIc, 75 parts diatomaceous earth, 3.5 parts wetting agent (calcium alkylbenzene sulfonate) and 3.5 parts dispersing agent (calcium lignin sulfonate) were thoroughly pulverized and mixed to give an 18% hydrated agent.

Manufacturing Example 4 Hydrated agent

Ten parts each of compounds Ia-Id, 10 parts each of compounds IIa-IIc, 55 parts diatomaceous earth, 20 parts white carbon, 3 parts wetting agent (sodium lauryl sulfate) and 2 parts dispersing agent (calcium lignin sulfonate) were thoroughly pulverized and mixed, to give a 20% hydrated agent.

Manufacturing Example 5 Hydrated agent

Ten parts each of compounds Ia-Id, 20 parts each of compounds IIa-IIc, 63 parts diatomaceous earth, 3.5 parts wetting agent (calcium alkylbenzene sulfonate) and 3.5 parts dispersing agent (calcium lignin sulfonate) were thoroughly pulverized and mixed, to give a 30%



hydrated agent.

Manufacturing Example 6                      Suspension agent

Five parts each of compounds Ia-Id, 20 parts each of compounds IIa-IIc, 3 parts polyoxymethylene [sic; polyoxyethylene] sorbitan monooleate, 3 parts CMC and 69 parts water were mixed and wet-pulverized until the particle size of the active ingredients was under 5  $\mu\text{m}$ , to give a 25% suspension agent.

Manufacturing Example 7                      Granular agent

One part each of compounds Ia-Id, 1 part each of compounds IIa-IIc, 1 part synthetic hydrated silicon oxide, 2 parts calcium lignin sulfonate, 30 parts bentonite and 65 parts kaolin clay were pulverized and mixed, respectively, followed by kneading thoroughly after adding water, granulation and drying, to give a 2% granular agent.

Manufacturing Example 8                      Emulsion agent

Five parts each of compounds Ia-Id, 15 parts each of compounds IIa-IIc, 14 parts polyoxyethylene styrylphenyl ether, 6 parts calcium dodecylbenzene sulfonate and 60 parts xylene were thoroughly mixed to give a 20% emulsion agent.

Next, the usefulness of the composition of the present invention as a microbicide is disclosed using test examples. Here, the compounds utilized in the experiments are shown in Tables 1 and 2 by compound symbols.

Also, the disease conditions of the test plants, i.e, the degrees of microbial plaque and disease spots on leaves are observed with the naked eye during the investigation to judge the disease prevention effects, which are calculated as the prevention value using the following method.



//insert a & b, p. 4//

- Key:
- |   |                                                                                                            |
|---|------------------------------------------------------------------------------------------------------------|
| 1 | Disease degree                                                                                             |
| 2 | a     Number of leaves of which more than 50% were found infected with microbial plaque and disease spots. |
|   | b     Number of leaves of which 25-50% were found infected with microbial plaque and disease spots.        |
|   | c     Number of leaves of which 10-25% were found infected with microbial plaque and disease spots.        |
|   | d     Number of leaves of which less than 10% were found infected with microbial plaque and disease spots. |
|   | e     Number of leaves of which none were found infected with microbial plaque and disease spots.          |
| 3 | Prevention value                                                                                           |
| 4 | Disease degree of the treated area                                                                         |
| 5 | Disease degree of the untreated area                                                                       |

#### Test Example 1

Experiment on the preventive effect for *Peronospora capsici* of cucumbers

Cucumber plants (variety: Sagami half-white) were planted in sandy soil in plastic pots.



These were cultivated for 14 days in a greenhouse, and young cucumber plants with young open leaves were obtained. Afterward, the hydrated agents according to Manufacturing Examples 4 and 5 were diluted with water to given concentrations and used as the test samples by spraying on the stems and leaves of the aforementioned young plants to insure that it adhered to all the leaves. After spraying, a fungal spore suspension of *Peronospora capsici* of cucumbers was sprayed and inoculated. After inoculation, they were placed in a humid place at 20°C for one day, followed by growth for 8 days under illumination. The disease conditions were then examined and the prevention values were calculated. Table 3 shows the results.

Table 3. Experiment on the preventive effect for *Peronospora capsici* of cucumbers

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//insert Table 3, p. 5//



- 2 Concentrations of active ingredients
- 3 Prevention value
- 4 Compound

### Experimental Example 2

Experiment on the preventive effect for *Peronospora capsici* of cucumbers

Cucumber plants (variety: Sagami half-white) were planted in sandy soil in plastic pots. These were cultivated for 14 days in a greenhouse, and young cucumber plants with young open leaves were obtained. A fungal spore suspension of *Peronospora capsici* of cucumbers was sprayed and inoculated on the young plants. After the inoculation, they were placed in a humid place at 20°C for one day and the hydrated agents according to Manufacturing Example 4 and 5 were diluted with water to given concentrations and used as the test samples by spraying on the stems and leaves to insure that it adhered to all the leaves. After the spraying, they were allowed to grow for 8 days under illumination. The disease conditions were then examined and the prevention values were calculated. Table 4 shows the results.



Table 4. Experiment on the preventive effect for *Peronospora capsici* of cucumber

//insert Table 4, p 5//

Key	1	Test compounds
	2	Concentrations of active ingredients
	3	Prevention value
	4	Compound ____



### Experimental Example 3

Experiment on the preventive effect for *Phytophthora infestans* of tomatoes

Sandy soil was packed in plastic pots and tomato (seeds) (Ponterosa [transliteration]) were planted and cultivated for 20 days to give young tomato plants having developed Nos. 2 to 4 leafy stems. The hydrated agents according to Manufacturing Example 4 and 5 were diluted with water to given concentrations and used as the test samples by spraying on the stems and leaves of the aforementioned young plants to insure that it adhered to all the leaves. After spraying, a fungal spore suspension of methalaxyl (a commercial microbicide) resistant *Phytophthora* was sprayed and inoculated. After inoculation, they were placed in a humid place at 20°C for one day, followed by growth for 5 days under illumination. The disease conditions were then examined and the prevention values were calculated. Table 5 shows the results.

Table 5. Experiment on the preventive effect for *Phytophthora infestans* of tomatoes

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//insert Table 5, p. 6//



Key	1	Test compounds
	2	Concentrations of active ingredients
	3	Prevention value
	4	Compound ____

#### Experimental Example 4

Experiment on the preventive effect for *Peronospora viticola* of grapes

Sandy soil was packed in plastic pots and grape (seeds) were planted and cultivated for 50 days in a greenhouse to give young grape plants having developed Nos. 3 to 4 leafy stems. A fungal spore suspension solution of *Peronospora viticola* of grapes was sprayed and inoculated on the young plants. After inoculation, they were placed in a humid place at 20°C for one day and the hydrated agents according to Manufacturing Example 4 and 5 were diluted with water to given concentrations and used as the test samples by spraying on the stems and leaves to insure that it adhered to all the leaves. After spraying, they were allowed to grow for 5 days under illumination. The disease conditions were then examined and the prevention values were calculated. Table 6 shows the results.



Table 6. Experiment on the preventive effect for *Peronospora viticola* of grapes

//insert Table 6//

Key	1	Test compounds
	2	Concentrations of active ingredients
	3	Prevention value
	4	Compound ____

Effect of the invention

The composition of the present invention shows extraordinary effects on phycomycetes



diseases in particular and is useful as an agriculture and garden microbicide.